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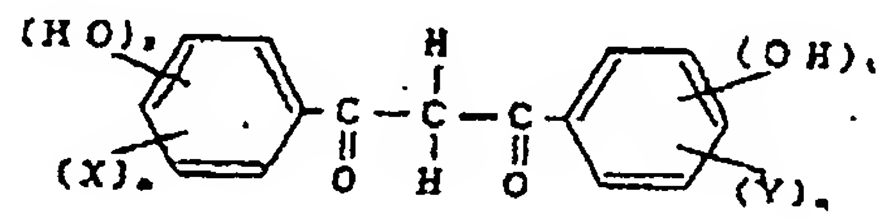
Applicant: Pierce Co. Ltd.

1. Title of the Invention

UV ABSORBER-INCORPORATED MICROCAPSULES, PROCESS FOR PRODUCING THE SAME AND COSMETIC MATERIALS CONTAINING THE MICROCAPSULES

2. Scope of the Claim for Patent

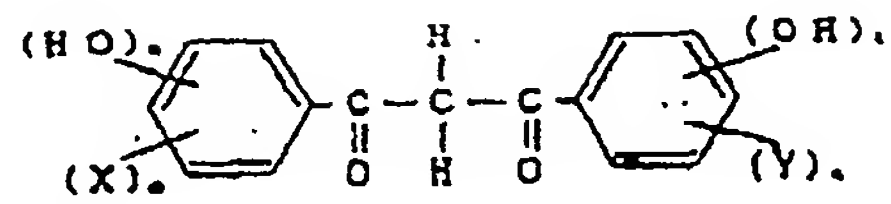
(1) UV absorber-incorporated microcapsules, in which a dibenzoylmethane derivative represented by the general formula:



(wherein m number of X and n number of Y each represents identical or different an alkyl group, an alkoxy group having from 1 to 18 carbon atoms, a carboxyl group or halogen, and m and n are each an integer of from 0 to 3, k + 1 is an integer of from 1 to 4) in fine spherical particles mainly comprising silica and having an average grain size of from 0.1 to 30 μm.

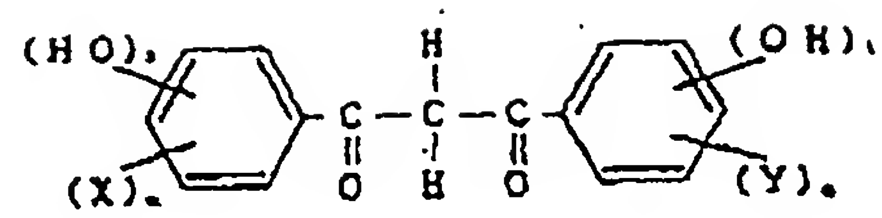
(2) A process for producing UV absorber-incorporated microcapsules, which comprises dissolving a

dibenzoylmethane derivative represented by the general formula:



(wherein m number of X and n number of Y each represents identical or different an alkyl group, an alkoxy group having from 1 to 18 carbon atoms, a carboxyl group or halogen, and m and n are each an integer of from 0 to 3, k + l is an integer of from 1 to 4) in an aqueous solution of alkali metal silicate, mixing the aqueous solution, water and an organic solvent having solubility relative to the dibenzoylmethane derivative of 5% or less to form a W/O type emulsion, and then mixing the emulsion with an aqueous acidic solution capable of forming water insoluble precipitation by neutralization reaction of the alkali metal silicate with an alkali soluble material of the dibenzoyl methane derivative, and then optionally, filtering, washing with water and drying the mixture to allow the dibenzoyl methane derivative to be incorporated in fine spherical particles mainly comprising silica and having an average grain size of from 0.1 to 30 μm.

(3) A cosmetic material containing microcapsules in which a dibenzoylmethane derivative represented by the general formula:



(wherein m number of X and n number of Y each represents identical or different an alkyl group, an alkoxy group having from 1 to 18 carbon atoms, a carboxyl group or halogen, and m and n are each an integer of from 0 to 3, k + 1 is an integer of from 1 to 4) is incorporated in fine spherical particles mainly comprising silica and having an average grain size of from 0.1 to 30 μm.

3. Detailed Description of the Invention

(Industrial Field of Use)

The present invention concerns microcapsules incorporated with a UV absorber, a process for producing the microcapsules, as well as it relates to cosmetic materials containing the microcapsules incorporated with the UV absorber.

[Prior Art]

As is well-known so far, UV radiation gives various effects on the skin.

Namely, UV rays in a UV-B region (290 to 320 nm) cause red spots or blisters on the skin, which blackens the skin after inflammation.

Although UV rays in a UV-A region (320 to 400 nm) scarcely cause red spots, it is the same in view of blackening the skin as in a case of UV-B region.

In addition, UV radiation causes pigmentation such as spots or freckles or causes aging or modification of skin.

Then, in order to solve such various kinds of problems, various UV absorbers have been developed and, and they are contained, for example, in cosmetic materials.

In particular, cosmetic materials are greatly required for screening UV radiation to the skin, and accordingly, those cosmetic materials have been put into practical use that prevent endermic absorption of UV rays by incorporating inorganic pigments such as titanium dioxide and zinc oxide capable of reflecting UV rays in addition to the UV absorbers described above (mainly those of organic series, for example, paraaminobenzoate derivatives, benzotriazole derivatives, benzophenone derivatives and cinnamate derivatives).

(Subject to be Solved by the Invention)

(i) However, in a case of cosmetic materials containing the organic type UV absorber, the compatibility of the UV absorber to a cosmetic base and feeling on use have not been always satisfactory.

In addition, there have been problems that when applying them to the skin, the UV absorber itself gives stimulation on the skin, and when such a UV absorber

absorbs light energy, it gives temporary stimulation to the skin.

In any of the cases, since cosmetic materials containing existent UV absorbers involve any of the problems described above, they are restricted to specific kinds when actually applied onto the skin.

(ii) On the other hand, in a case of cosmetic materials containing the inorganic pigment described above, they result in problems of stimulation to the skin, but since they are not contained in cosmetic materials with an aim of a UV absorbing effect, they involve drawback that they can not screen UV radiation sufficiently, and can not prevent epidermal absorption of UV rays.

In particular, cosmetic materials containing an inorganic pigment of grain size of shielding visible rays (relatively large grain size) less absorb radiation in a UV region.

On the other hand, when blending fine titanium dioxide particles, UV shielding effect can be obtained by the light scattering effect, but they provide conspicuously white skin by the scattering of light. In addition, since they are poor in adhesion to the skin, and lack in spreadability, they adhere thick on the skin, resulting in make-up with no transparency.

In any of the cases, there have scarcely been

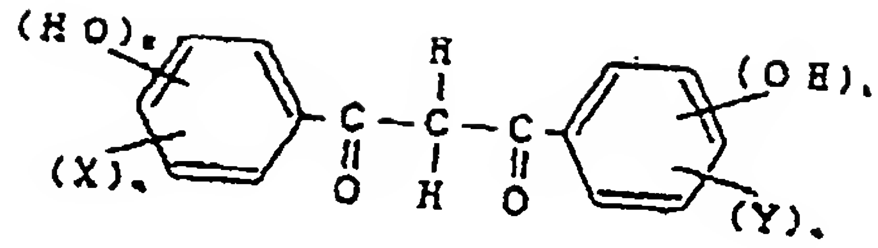
developed cosmetic materials which prevent epidermal UV absorption and satisfy various conditions such as safety and compatibility to the skin.

The present invention has been accomplished for solving all the problems described above, and the object thereof is to provide cosmetic materials capable of preventing epidermal UV absorption by satisfactory absorption and screening effects of UV radiation harmful to the skin, and excellent in safety, adhesion to the skin and spreadability, with excellent feeling on use and transparency.

(Means for Solving the Subject)

The present inventors have made an earnest study for solving such problems, and as a result, they found that when a certain kind of organic UV absorber is incorporated in a certain kind of fine inorganic spherical particles to form microcapsules, and they are blended to cosmetic materials, all of the problems can be solved, and have accomplished the invention.

Namely, the invention provides a UV absorber-incorporated microcapsules, and a process for producing the microcapsules, and cosmetic materials containing the microcapsules, wherein the microcapsules have a feature of incorporating dibenzoylmethane derivative represented by the general formula:



(wherein m number of X and n number of Y represent identical or different an alkyl group, an alkoxy group having from 1 to 18 carbon atoms, a carboxyl group, or halogen, and m and n are each an integer of from 0 to 3, k + l is an integer of from 1 to 4) is incorporated in fine spherical particles mainly comprising silica and having an average grain size of from 0.1 to 30 μm.

In addition, the process for producing the microcapsules comprises dissolving the dibenzoylmethane derivative represented by the chemical formula described above in an aqueous solution of alkali metal silicate, mixing the aqueous solution, water and an organic solvent having solubility relative to the dibenzoylmethane derivative of 5% or lower to form a W/O emulsion, and then mixing the emulsion with an aqueous acidic solution capable of forming water insoluble precipitation by neutralization reaction of the alkali metal silicate with an alkali soluble material of the dibenzoyl methane derivative, and then optionally, filtering, washing with water and drying the mixture to allow the dibenzoyl methane derivative to be incorporated in fine spherical particles mainly comprising silica and having an average grain size of

from 0.1 to 30 μm.

Further, the feature of the present invention as a cosmetic material is incorporation of microcapsules as described above.

The dibenzoylmethane derivative represented by the general formula includes, for example, the following compounds.

(i) 4,4'-dihydroxydibenzoylmethane,

(ii) 4-hydroxy-4'-methoxydibenzoylmethane,

(iii) 2,4'-dihydroxydibenzoylmethane,

(iv) 2,4-dihydroxy-4'-methoxydibenzoylmethane,

(v) 2,4-dihydroxydibenzoylmethane,

(vi) 4-hydroxydibenzoylmethane,

(vii) 2,4,4'-trihydroxydibenzoylmethane,

(viii) 4-hydroxy-4'-methyldibenzoylmethane,

(viiii) 2,4-dihydroxy-4'-chlordibenzoylmethane,

(x) 4-hydroxy-4'-chlordibenzoylmethane,

(xi) 2,4'-dihydroxy-4-methoxydibenzoylmethane, and

(xii) 4-hydroxy-3'-carboxydibenzoylmethane,

However, the kinds of the dibenzoylmethane derivatives in the present invention are not limited to (i) to (xii) described above.

In addition, in the production process of the microcapsules, the alkali metal silicate can include, for example, JIS No. 1 sodium silicate, JIS No. 2 sodium

silicate, JIS No.3 sodium silicate, sodium metasilicate, potassium silicate ($K_2O \cdot nSiO_2$, n=2 to 3.8).

An organic solvent to be used for the production process described above can include, for example, saturated aliphatic hydrocarbon such as n-hexane, decane and octane or aromatic hydrocarbon such as toluene, benzene, xylene, and in addition, alicyclic hydrocarbon such as cyclohexane.

Each of the solvents can be used of course alone or in combination of two or more of them.

In addition, as the emulsifier to be used for the production process, nonionic surfactants having an HLB value within a range from 3.5 to 6.0 are preferably used. The typical example can include sorbitane sesquioleate, sorbitane monooleate and polyoxyethylene sorbitane trioleate.

The aqueous acidic solution to be used in the production process can preferably include polyvalent anions such as sulfuric ions, and phosphoric ions. For example, when the dibenzoylmethane derivative is 4,4-dihydroxydibenzoylmethane or 4-dihydroxydibenzoylmethane, an aqueous acidic solution having a pH around 6 after the completion of the reaction is preferred, and the concentration is preferably about 1.5 mol/liter.

The kinds of the alkali metal silicate, the

organic solvent, the emulsifier and the aqueous acidic solution are not limited to those described above.

[Function]

(1) Concerning formulation mechanism

Fig. 1 is an explanatory view showing the process for forming steps of processing fine particles of microcapsules of the present invention.

At first, when a dibenzoylmethane derivative is dissolved in an aqueous solution of alkali metal silicate and the aqueous solution is mixed with an organic solvent, a W/O type emulsion comprising a mixed liquid of the dibenzoylmethane derivative and an aqueous solution of the alkali metal silicate as dispersion medium 1 and the organic solvent as a dispersion medium 2 is prepared as shown in Fig. 1(i).

Next, the emulsion is mixed with the aqueous acidic solution.

In this case, the following chemical reaction is taken place on the boundary of the dispersion medium 1 and the aqueous acidic solution.

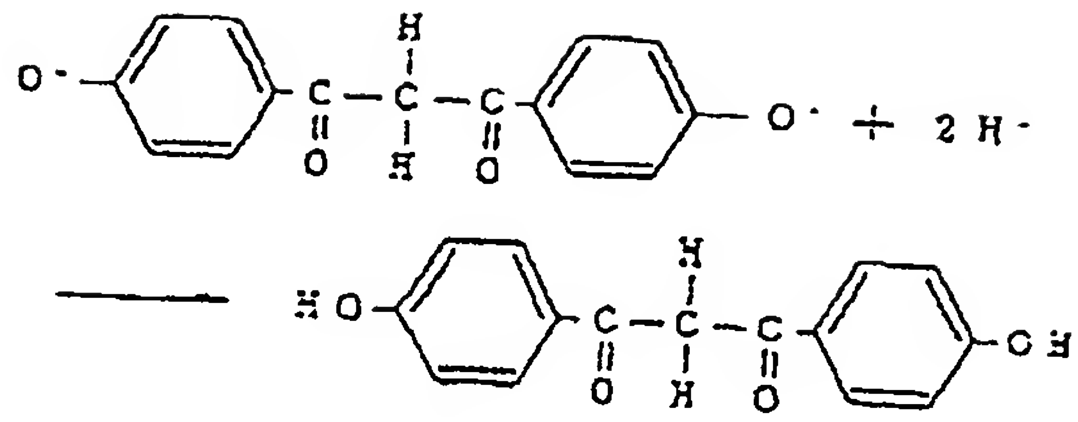
(a) $SiO_3^{-2} + 2H^+ \rightarrow SiO_2 + H_2O$

$(Si_2O_5^{2-} + 2H^+ \rightarrow 2SiO_2 + H_2O)$

and

(b) $R\text{-}O^- + H^+ \rightarrow R\text{-}OH$

(wherein R represents dibenzoylmethane skeleton)



(A)

In the reaction of (b), when R represents 4,4'-dihydroxydibenzoylmethane, the reaction proceeds as [A].

The chemical reaction on the boundary in the present invention proceeds as in the reaction formula, which is a coprecipitation reaction in which the two chemical reactions proceed simultaneously.

However, since the reaction of (a) proceeds more rapid than the reaction (b), a thin film 3 of silica is formed at first on the boundary, and the reaction on the boundary proceeds to the inside of an inner aqueous phase with lapse of time, so that microcapsules 5 are formed in a state where a reaction product 4 of the dibenzoylmethane derivative is incorporated in silica.

(2) Prevention of leaching of incorporated dibenzoylmethane derivative

Since the microcapsules incorporated with dibenzoyl methane derivative in fine spherical particles mainly comprising silica obtained by the reaction mechanism contain a large quantity of deposited water, they are preferably dried at a temperature as high as

100°C or higher at such an extent as not to modify the dibenzoylmethane derivative. By the procedures, leaching of the incorporated dibenzoylmethane derivative can be minimized.

Then, known surface treatment such as a treatment with silicone oil can optionally be conducted, so that the leaching of incorporated dibenzoylmethane derivative can be prevented.

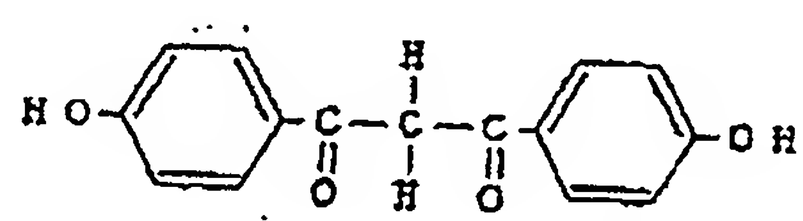
[Example]

Explanation will be made to examples of the present invention.

[Example of microcapsules]

Example 1

This example is an embodiment for microcapsules incorporated with 4,4'-dihydroxydibenzoylmethane as one example of the dibenzoylmethane derivative in the present invention.



[I]

Namely, microcapsules in the invention comprise 16.52% by weight of 4,4'-dihydroxydibenzoylmethane represented by the formula [I] incorporated in fine spherical particles mainly comprising silica and having an average grain size of 1.2 μm.

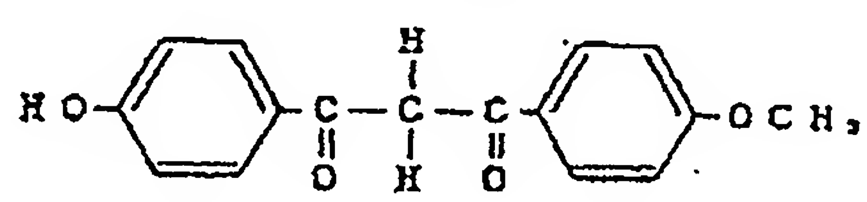
The scanning electron microscopic photographs of

the microcapsules of the invention are shown in the reference photographs in appended sheets. As the scanning electron microscope, JSM-T220A manufactured by Nippon Electron Co. Ltd. was used, and photographs were taken under acceleration voltage of 30 KV and magnifying factor of 10000 times.

Example 2

This example is an embodiment for microcapsules incorporated with 4-hydroxy-4'-methoxydibenzoylmethane as one example of dibenzoylmethane derivative.

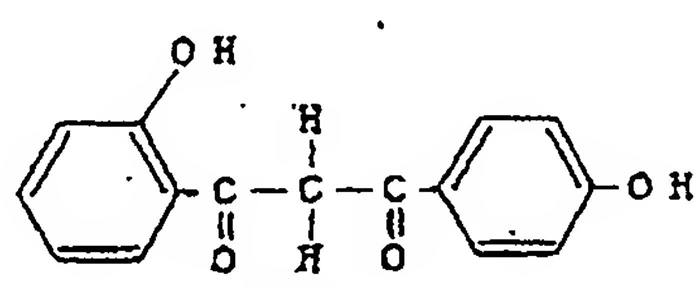
Namely, microcapsules in the invention comprise 18.93% by weight of 4-hydroxy-4'-methoxydibenzoylmethane represented by the formula [II] incorporated in fine spherical particles mainly comprising silica and having an average grain size of 1.3 µm.



(II)

Example 3

This example is an embodiment for microcapsules incorporated with 2, 4-dihydroxydibenzoylmethane as one example of the dibenzoylmethane derivative according to the invention.



(III)

Namely, microcapsules in the invention comprise

22.93% by weight of 2,4'-dihydroxydibenzoylmethane represented by the formula [III] in fine spherical particles mainly comprising silica and having an average grain size of 1.7 μm.

In addition, the grain size of the microcapsules of the invention is not limited to each of the examples, and, in short, those of fine spherical particles constituting the outer wall of the microcapsules having an average grain size of from 0.1 to 30 μm may suffice.

Reference Example

UV absorption spectra were measured for the microcapsules of Examples 1 to 3, and tests for slipping friction were conducted.

(1) UV absorption spectrum

A powder of the specimen of each example was added by 10% by weight in an official white Vaseline and kneaded sufficiently to disperse, and coated between quartz plates to a thickness of 15 μm, and UV spectrum was measured.

As a result, as shown in Fig. 2, microcapsules of each of examples showed absorption spectrum corresponding to that of the incorporated dibenzoylmethane derivative, and showed sufficient absorbability relative to UV rays which result in problems in view of the relation with endermic

absorption.

(2) sliding friction

The specimen of each of examples was coated thinly on a polished plate glass, and a flat glass plate on which a 200 g weight was loaded was placed thereon. When it was drawn horizontally by a spring scale, the tensile load upon sliding was measured and the results were shown by bar graph as in Fig. 3.

As a result, it was found that the sliding friction was small compared with that of talc, sericite, iron oxide red and titanium oxide contained in ordinary cosmetics.

This is considered to be attributable to the fact that since the microcapsules are true spheres, they are satisfactory in rolling effect (rolling property) compared with that of talc and the like.

[Example of process for producing microcapsules]

Example 4

This example is an embodiment of a process for producing microcapsules incorporated with 4,4-dihydroxybenzoylmethane described in Example 1.

At first, 6.0g of 4,4'-dihydroxydibenzoylmethane was dissolved in 250 mol solution of No. 1 sodium silicate of 0.8 mol/liter, and the aqueous solution was poured to

400 mol of 5% toluene solution of a mixture of sorbitan sesquioleate and polyoxyethylene sorbitan trioleate (mixing ratio 4/1) and emulsified for 5 min by a homomixer to prepare a W/O type emulsion.

Then, the emulsion was poured in 1500 ml of an aqueous mixed solution of 0.6 mol/liter of ammonium sulfate,

0.68 mol/liter of dihydrogen sodium phosphate and 0.12 mol/liter of hydrogen disodium phosphate, stirred for 1 hour, and stood still for one night. Then, it was separated into a solid and a liquid by centrifugation, and then filtered, washed with water and dried at 150°C.

With such procedures, 28.8g of microcapsules having an average grain size of 1.2 µm incorporated with 4,4'-dihydroxydibenzoylmethane in fine spherical particles mainly comprising silica were obtained.

Example 5

This example is an embodiment of a process for producing microcapsules incorporated with 4-hydroxy-4'methoxydibenzoylmethane described in Example 2.

At first, 5.0g of 4-hydroxy-4'-methoxydibenzoylmethane was dissolved in 200 mol solution of 0.75 mol/liter of No. 1 sodium silicate, and the aqueous solution was poured to

350 ml of 6% n-hexane solution of a mixture of sorbitan

sesquioleate and polyoxyethylene sorbitan trioleate (mixing ratio 3:1) and emulsified for 5 min by a homomixer to prepare a W/O type emulsion.

Then, the emulsion was poured in 1200 ml of an aqueous mixed solution of 1.25 mol/liter of dihydrogen sodium phosphate and 0.25 mol/liter of hydrogen disodium phosphate, stirred for 1 hour, and then the same procedures as in Example 4 were conducted to obtain 22.8 g of microcapsules having an average grain size of 1.3 μm and incorporated with 18.93% by weight of 4-hydroxy-4'-methoxydibenzoylmethane in fine spherical particles mainly comprising silica.

Example 6

This example is an embodiment of a process for producing microcapsules incorporated with 2,4'-dihydroxybenzoylmethane described in Example 3.

At first, 8.0g of 2,4'-dihydroxybenzoylmethane was dissolved in 250 mol solution of 0.8 mol/liter of No. 1 sodium silicate, and the aqueous solution was poured to 400 mol of 5% toluene solution of a mixture of sorbitan monooleate and polyoxyethylene sorbitan monooleate (mixing ratio 6:1) and emulsified for 5 min by a homomixer to obtain a W/O type emulsion.

Then, the emulsion was poured in 1500 ml of an aqueous mixed solution of 0.6 mol/liter of ammonium

sulfate, 0.70 mol/liter of hydrogen disodium phosphate and 0.1 mol/l of hydrogen disodium phosphate, stirred for 1 hour, and the same procedures as those in Example 4 were conducted to obtain 30.8 g of microcapsules having an average grain size of 1.7 µm incorporated with 2,4'-dihydroxydibenzoylmethane in fine spherical particles mainly comprising silica.

[Examples of cosmetic materials]

Microcapsules described above can be blended in cosmetic materials and pharmaceutical products for the purpose of preventing undesired effects of UV radiation applied on the skin.

Although the blending amount of the microcapsules of the invention to the cosmetic material is different depending on the kinds of the cosmetic materials, it is generally preferably from 0.1 to 20% by weight and, especially, from 0.5 to 10% by weight as the dibenzoylmethane derivative.

The cosmetic materials of the invention are prepared by blending the microcapsules in known cosmetic bases by an ordinary method, and formulating them into various shapes of agents including creams, solutions, sticks, milky lotions, foundations and ointments.

Namely, it is possible to produce all sorts of cosmetic materials having UV absorbing effect, including

cosmetic oils of oil bases, oily creams blended with a large quantity of an oil base, oily milky lotions, weakly oily creams and weakly oily milky lotion, basic cosmetic products such as water-based beauty solution to various make-up cosmetic materials such as oil-based foundations and lipsticks by selectively using the microcapsnles in accordance with cosmetic bases.

Next, examples of the cosmetic materials of the invention will be explained.

Example 7

This example is an embodiment of cosmetic materials in which the microcapsules of the invention are blended in a so called powder foundation.

The composition of the cosmetic material of the invention is as follows.

	Ingredients	% by weight
(1)	microcapsules of Example 1	25.0
(2)	talc	balance
(3)	mica	30.0
(4)	titanium mica	1.0
(5)	titanium oxide	8.0
(6)	iron oxide red	0.7
(7)	yellow iron oxide	1.8
(8)	black iron oxide	0.2
(9)	crystalline cellulose	0.2

(10)	methylpolysiloxane	4.0
(11)	liquid paraffin	3.0
(12)	squalane	4.0
(13)	perfume	q.v.
(14)	antiseptic agent, antioxidant	trace amount

When producing the cosmetic material of the invention, (1) to (9) are mixed sufficiently by a Henschel mixer, and a mixture of other ingredients was added homogeneously, processed by a pulverizer followed by press-molding.

The cosmetic material of the invention had transparency and adhesion, and could be adhered on skin thinly and uniformly.

It was confirmed that it had an extremely high UV screening effect.

In addition, it provided no conspicuous whiteness as in cosmetic materials blended with existent inorganic pigments.

Further, it could provide soft feeling on use, with satisfactory cosmetic retainability.

Example 8

This example is an embodiment for a cosmetic material in which the microcapsules of the invention are blended in an oily foundation.

The cosmetic composition of the cosmetic material

of the example is as follows.

	Ingredients	% by weight
(1)	bees wax	11.0
(2)	ceresin	1.0
(3)	Vaselin	3.0
(4)	purcellin oil	28.0
(5)	squalane	2.0
(6)	isostearic acid	4.0
(7)	titanium oxide	28.0
(8)	titanium mica	4.0
(9)	talc	5.0
(10)	iron oxide red	1.2
(11)	yellow iron oxide	3.1
(12)	black iron oxide	0.3
(13)	microcapsules of Example 2	10.0
(14)	perfume	q.v.
(15)	antiseptic agent, antioxidant	trace amount

When producing the cosmetic material of the invention, from (7) to (13) were added to a portion of (4), and processed with a roller (a pigment portion), other ingredients were mixed, melted while heating, and the pigment portion was added and dispersed uniformly by a homomixer. After the dispersion, it was cooled to 50°C while stirring.

The cosmetic material of the invention had a high

UV screening effect with excellent feeling on use and transparency.

In addition, it was excellent in prevention of conspicuous whiteness, with satisfactory cosmetic retainability.

Example 9

This example is an embodiment of a cosmetic material in which the microcapsules of the invention are blended in a W/O type cream.

The composition of the cosmetic material of the embodiment is as follows.

Ingredient	% by weight
(1) solid paraffin	5.0
(2) bees wax	5.0
(3) microcrystalline wax	10.0
(4) Vaselin	10.0
(5) squalane	35.0
(6) polyoxyethylene (20) sorbitan monolaurate	1.0
(7) sorbitan sesquioleate	5.0
(8) microcapsules in Example 1	10.0
(9) purified water	balance
(10) perfume	q.v.
(11) antiseptic agent, antioxidant	trace amount

When producing the cosmetic material, (8) was at

first added to (9), heated and kept at 80°C (an aqueous phase), mixed with other ingredients, melted while heating and kept at 80°C (an oil phase). (8) of the aqueous phase was dispersed uniformly by a homomixer to emulsify, the aqueous phase was added to the oil phase, and emulsified uniformly by a homomixer, and kneaded while cooling after the emulsification to obtain the cosmetic material.

The cosmetic material of the invention was excellent in a UV screening effect, and was more safely compared with existent cosmetic materials.

In addition, it adheres on skin thinly uniformly with excellent feeling on use and transparency.

Further, it was excellent in prevention of conspicuous whiteness with satisfactory cosmetic retainability.

Example 10

This example is an embodiment for a cosmetic material in which the microcapsules of the invention are blended in an O/W type cream.

The composition of the cosmetic material of the embodiment is as follows.

	Ingredients	% by weight
(1)	bees wax	10.0
(2)	stearyl alcohol	5.0

(3)	hydrated lanolin	8.0
(4)	squalane	33.0
(5)	glycerin monostearate	2.0
(6)	polyoxyethylene(20)sorbitan monolaurate	2.0
(7)	propylene glycol	5.0
(8)	microcapsules of Example 2	10.0
(9)	purified water	balance
(10)	perfume	q.v.
(11)	antiseptic agent, antioxidant	trance amount

When producing the cosmetic material of the example, (7) and (8) were added to (9), heated and kept at 70°C (aqueous phase). Other ingredients were mixed, melted while heating and kept at 70°C (oil phase). (8) in the aqueous phase were dispersed uniformly by a homomixer, and then the oil phase was added to the aqueous phase, emulsified homogeneously by a homomixer, stirred while cooling after the emulsification.

The cosmetic material of the example is excellent in a UV screening effect and is higher in transparency compared with existent cosmetic material.

In addition, it adheres on skin thinly uniformly with excellent feeling on use and transparency.

In addition, it was excellent in prevention of conspicuous whiteness, with satisfactory cosmetic

retainability.

Example 11

This example is an embodiment of a cosmetic material in which the microcapsules of the invention are blended in a lip cream.

The composition of the cosmetic material of the embodiment is as follows.

Ingredients	% by weight
(1) microcapsules of Example 1	8.0
(2) candellila wax	3.0
(3) ceresin	15.0
(4) resiner	5.0
(5) octyl dodecanol	7.0
(6) diisostearylate	35.0
(7) glycerin tri-2-ethylhexanoate	22.0
(8) neopentyl glycol dioctanoate	4.3
(9) perfume	q.v.
(10) antiseptic agent, antioxidant	trace amount

When producing the cosmetic material of the example, (1) was added to a portion of (6), processed with three rollers to form a pigment portion. Next, other ingredients were mixed, melted while heating, and then the pigment portion was added, and dispersed homogeneously by a homomixer. The material after the dispersion was poured into a mold and rapidly cooled to

form a stick, and put into a container and subjected to framing.

The cosmetic material of the example is excellent in a UV screening effect and has higher safety than existent cosmetic materials.

In addition, it adheres thinly and uniformly on lips, with excellent feeling on use and transparency.

(Effect of the Invention)

(i) As described above, since the microcapsules of the invention are prepared by incorporating a dibenzoylmethane derivative as a UV absorber in fine spherical particles having an average grain size of from 0.1 to 30 μm and mainly comprising silica, and the UV absorber is not in direct contact with skin, stimulation on skin is mitigated, so that they have a remarkable effect that the safety is greatly enhanced compared with that of existent UV absorbers.

Accordingly, cosmetic materials containing such microcapsules have an advantage capable of obtaining an effect of mitigating stimulation on skin.

(ii) They also provide an effect that since microcapsules pre se are a powder, they can be blended easily in a cosmetic base for which the blending of existent UV absorbers has been difficult.

(iii) In addition, since the microcapsules are a true

spherical powder, spreadability of cosmetic materials containing such microcapsules is extremely satisfactory compared with that of existent cosmetic materials containing inorganic pigments, so that they have an effect that they adhere on skin thinly and uniformly with no thick adhesion, this applies no burden on skin.

(iv) In addition, in such a cosmetic material, since the fine spherical particles constituting the outer wall of the microcapsules comprise silica having a refractive index substantially equal with that of an oil solvent in the cosmetic material base, they have an advantage not providing impression of conspicuous whiteness of a face due to the scattering of light, as in a cosmetic material containing existent titanium oxide.

(v) The cosmetic materials described above have advantage of excellent transparency, excellent press-castability and cosmetic retainability.

(vi) Further, since the production process for producing microcapsules of the invention comprises dissolving a dibenozylmethane derivative in an aqueous solution of an alkali metal silicate, mixing the aqueous solution with an organic solvent to form a W/O emulsion, and then mixing an aqueous acidic solution capable of forming water insoluble precipitate by neutralization reaction of the alkali metal silicate with an alkali solubilized

product of a dibenzoylmethane derivative, the dibenozylmethane derivative is incorporated in the fine spherical particles mainly comprising silica.

In particular, in the following co-precipitation reaction on the boundary:

(a) $SiO_3^{2-}+2H^+ \rightarrow SiO_2 + H_2O$

($Si_2O_3^{2-}+2H^+ \rightarrow 2\ SiO_2 + H_2O$)

and

(b) $R - O^- + H^+ \rightarrow R{-}OH$

(wherein R represents a dibenzoylmethane skeleton), the reaction of (a) proceeds more rapid than the reaction of (b), the dibenzoylmethane derivative is reliably incorporated in the fine spherical particles, and accordingly, it can provide an effect that production of microcapsules can be conducted reliably.

4. Brief Description of the Drawings

Fig. 1 is an explanatory view showing a production process of forming the microcapsules,

Fig. 2 is a chart of UV absorption spectrum of the microcapsules of one example,

Fig. 3 is a graph of a sliding friction test of the microcapsules of one example.

Fig. 1

(i), (ii), (iii)

Fig. 2:

Absorption, wavelength (nm)

A: microcapsules of Example 1

B: microcapsules of Example 2

C: microcapsules of Example 3

D: fine silica spherical particles (average grain size of 1.2 μm),

E: talc

Fig. 3

Tensile load (g),

Nylon powder,

Iron oxide red

Titanium oxide

Sericite,

Talc

A: microcapsules of Example 1

B: microcapsules of Example 2

C: microcapsules of Example 3

Voluntary amendment June 22, 1990

The Director of the Patent Office Fumio Yoshida

1. Indication of the Case

   Patent Application No. 72234/1989

2. Title of the Invention

   UV absorber-incorporated microcapsules, process for producing the same and cosmetic materials containing the microcapsules

3. Person for amendment

   Relation with the case Applicant

   Residence

   Name Pierce Co. Ltd.

4. Proxy

   Residence

   Zip code: 542

   Osaka-city, Chuo-ku, Minami Senba 2-5-8

   Nagahori Community Building

   Tel No. 06-271-7908

   Name (7433) Patent Attorney Noboru Fujimoto

5. Date of instruction for amendment Voluntary

6. Number of inventions increased by the amendment

7. Object of the amendment

   Column of detailed description of the invention in